

Rocky Flats Environmental Technology Site

Actinide Migration Evaluation

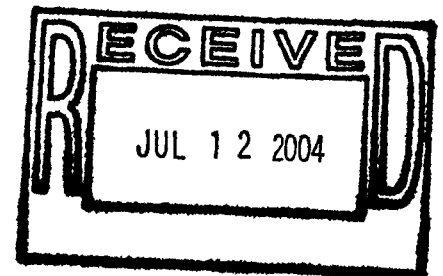
Meetings October 5 & 6, 1999

Advisory Group

Greg Choppin, David Clark, David Janecky, Leonard Lane, Kirk Nordstrom

Summary and recommendations for path forward

The integration of Site activities and knowledge is improving and being engaged in decision and planning processes. Simultaneously, there is only a weak connection with the rest of the DOE Complex and the international radionuclide remediation community. The Advisory Group recognizes that part of its charge is to bring an external perspective to the Site. However, direct interaction with the national and international communities needs to be broader (e.g., not participating in the recent Actinide Migration99 was a missed opportunity for Site leaders). Similarly, the Advisory Group will be encouraging AME projects to include comparative information from the literature and other sites in their reports. Putting results and conclusions in context is particularly important for integrated risk and cleanup evaluations where the detailed chemical and physical behavior of actinides has both subtle and major impacts (e.g. limitation of RESRAD models to soluble transport and partitioning models, as compared to observed particulate transport of plutonium and americium).



ADMIN RECORD

Results and Discussions

Erosion Model -- Chromec and Wetherbee

Although the report of erosion modeling activities was very brief and only summarized the highlights, it was evident that a great deal of time and effort has gone into calibrating the WEPP Hillslope Model to use in actinide migration predictions. The next obvious task is to calibrate WEPP, using the recently acquired the rainfall simulator data, to derive improved estimates of soil erodibility parameters (K_i , K_r , and TAU_c) and effective hydraulic conductivity, K_e . The committee was assured this work would be accomplished soon.

The advisors pointed out that the stream channel water and sediment routing and the WEPP hillslope modeling are not independent tasks but are interrelated. Parameter estimates required for the channel routing will depend upon input from the hillslopes and total sediment yield at the watershed outlet can, in part, be used to help evaluate WEPP calculations on the hillslopes.

Finally, as the Actinide Migration studies are heavily dependent on erosion and sediment transport modeling, we felt it appropriate to make a brief statement on modeling and user applications and interpretations. Erosion simulation models are developed to assist in evaluating soil detachment, sediment transport and deposition. In cases where significance is placed on results of the analyses, the user should have independent methods for checking the accuracy of the outcomes. The use of independent methods to check the modeling results is a key part of any modeling study and should be planned, conducted and documented with the same thoroughness as every other step in the modeling and analyses. As in all applications of science and technology, proper use of simulation models, their input data, and interpretation of the resulting model output are the responsibility of the user.

Geochemical Modeling of Uranium at the Solar Ponds Plume -- Ball

Some delays have been experienced in acquiring the ground-water analyses for the Solar Ponds Plume (SPP) and in screening them for reliability. From many hundreds of analyses, a small subset have been found to be sufficiently complete and with good charge balance to speciate and test for solubility controls.

The results are being organized in 2 phases. The first phase, which is 90% complete, reviews the general hydrogeochemical characteristics of the SPP ground waters, describes the program used for geochemical equilibrium calculations (PHREEQC), tabulates and references the thermodynamic database used for uranium and auxiliary compounds and aqueous species, presents the main uranium species in the ground water, and portrays likely mineral solubilities with plots of saturation indices and a sensitivity analysis.

The results indicate that for uranium minerals, all are undersaturated. None appear to reach equilibrium solubility although some reach to about an order of magnitude of the equilibrium value. If it is confirmed that no mineral solubility control can be clearly delineated, the apparent attenuation of uranium in the plume relative to the plume of nitrate is likely caused by adsorption.

The second phase of this work will apply mass balance modeling to the ground waters, to quantify the amounts of minerals dissolved and precipitated during flow and to see how these natural processes are affected by the presence of the pollutants in the plume. Any further attempts to model the ground-water chemistry must be based on such an analysis. A sensitivity analysis will also be performed on these results.

Overview of RESDAD model -- Roberts

There still seems to be a failure in the application of the Resrad model to understand the limitations of K_D values. K_D values represent a single interaction parameter. If we are dealing with a static system, such a measurement has validity in modeling the solubility of a species. However, in migration in the medium, the use of a K_D requires a measurement of the number of theoretical plates (i.e., interactions) over the plume. For example, in movement down a thin column of material, it is not uncommon that actinide species will encounter 1000 plates over perhaps a one inch path length. Therefore, the K_D would have to be increased by distribution, which accounts for the dynamic conditions in a complex pore space. The use of the single K_D value representing an averaged interaction from a static batch measurement is completely unacceptable. It is unscientific and it ignores the proper use of K_D in modeling of migration through a column system. Of course, this also ignores the fact that actinide elements will have varying degrees of interaction with different kinds of soil components. Even the static K_D measured in one system, may not be applicable to a static measurement in another system of different soil.

Finally, we note that a major concern, with respect to the RESRAD model, is that it is a solubility based model and the dominant process at Rocky Flats is colloidal transport.

Kds -- Chromec

The discussion on an initial compilation of K_D for soils is a promising approach and should be continued by Kaiser Hill. In fact, it should be expanded to include K_D measurements from Europe and Japan in connection with their repository and environmental studies. Russia particularly has been doing a great deal in this area for the Chernobyl area, as well as in planned remediation of some of the highly contaminated nuclear sites.

Update on work on soil aggregating properties -- Ranville

Dr. Ranville, Colorado School of Mines, provided an update on his investigations of soil properties at RFETS funded by the EPA. He reviewed his objective and previous results. The objective is to determine the influence of soil size and aggregation properties on the mobility of plutonium in a small drainage between the SID and the 903 pad. The objective is divided into 3 categories: (1) to determine the Pu inventory as a function of size fraction and the mass balance for the amount of Pu for each fraction, (2) to determine the degree of transport for each size fraction, and (3) to determine the aggregate strength and main binding agent(s).

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The Pu resides in the top portion of the soil (top few centimeters) and occurs at higher concentration in the smallest size fraction (< 10 micrometers, 50-60 pCi/g for sample 5298), consistent with the results of other investigators. However, most of the mass of Pu resides in the larger size fraction (> 10 micrometers) because the larger size fraction makes up most of the soil (97% for sample 5298).

Five methods for investigating disaggregation are being used: (1) deionized water, (2) ultrasound, (3) hexametaphosphate (a detergent), (4) hydrogen peroxide, and (5) citrate-dithionate-bicarbonate. Each of these solutions selectively destroys a different component of the soil. Following each disaggregation, the soil is then size separated and analyzed for surface area, morphology, chemical composition, and Pu and Am. Organic carbon is also determined in the leach solutions as well as in the original soil. Redox agents, such as hydrogen peroxide, are only partly selective in destroying components of soils and can affect the oxidation states of iron, plutonium, etc. and some attention should be paid to its possible perturbation of the distribution of plutonium due to redox interactions.

The organic carbon content of the soils tends to vary little from the top of the watershed to the bottom (3-4%). The clay fraction (< 2 micrometers) also varies little (4-6%) and increases with degree of disaggregation. With sonication the clay content increases to 8.5-12%, and with hydrogen peroxide the range increases (6-20%) but the average value changes little. The redistribution of all size fractions changes significantly with degree of disaggregation. There is considerably more disaggregation (increase in smallest size fractions and decrease in largest size fractions) with the use of hexametaphosphate and hydrogen peroxide. This result suggests that organic matter could be a major source of soil binding at RFETS.

Remaining analyses and Pu and Am contents for the different size fractions and disaggregation categories have not been completed. Final results and a report are due at the end of the calendar year.

Update on Air Modeling -- Radian

The presentations on the air modeling were well done and indicated a good understanding on the part of the speakers about the model. The presentations were relatively straight forward and discussed the principle issues without getting sidetracked into details. As with the erosion model, the modeling results over estimated by a significant factor (10-100) when compared to actual measured values. The presenters discuss this discrepancy and indicated an understanding that could lead to this. Unlike the presentation of the RESRAD model in which there seem to be a desire to justify the modeling results and to use them even though they disagree with experiment because they were "conservative", the air modeling approach was more dedicated to evaluating the model to provide better agreement between measurement and modeling. The interpretation of the factors that could lead to such overestimation gave confidence in their understanding of the model, its factors and of the aspects of the site that could lead to present parameters in the model to an overestimation.

The approach that was suggested to divide the soil into fractions, and to model the components and to attach actinides based on the properties of the different fractions may lead to a useful model. They noted that a fire scenario methodology had been prepared but no modeling has been done using it this year. It is planned to study such fire scenarios in the future. In considering the two orders of magnitude disagreement with experimental data the factors that could be responsible involve dilution of surface soil compared to the underlying soil actinide levels, plume depletion and failure to account for the differences in particle sizes. Overall, the presentation was thorough and we have confidence that the team involved in the air modeling is producing a useful model that will approach reality in terms of matching experimental data.

Actinides in Concrete – Pat Ervin

An introduction to the approaches to handling and disposal of concrete present on the Rocky Flats site was presented. The volume of potentially contaminated concrete on the site is estimated to be hundreds of cubic meters. This material must be either determined to be stable and below release limits, so that it can be left on site, or it must be removed and disposed of during D&D activities. A critical question to be evaluated is what is the chemical state of the contaminants and how will those contaminants in recycled concrete behave in the geologic and soil environment. The suggestion to obtain some concrete samples to attempt to define better the speciation of the plutonium contamination is a good one. Better understanding can lead to the use of better techniques for handling and could result in substantial economic value.

Documents provided to advisory group

Figure 8a – 100 year average erosion map, Woman Creek western tile
Figure 8b – 100 year average erosion map, Woman Creek eastern tile
Figure 9 – 100 year average erosion map, south interceptor ditch (sid)
Figure 10 – 100 year average erosion map, Walnut Creek
Viewgraphs for RESRAD Overview
RESRAD v5.0 manual front pages and Appendix E Water Pathway Factors,
Appendix H , and Appendix J
Viewgraphs for air transport and deposition of actinides
Air transport and deposition of actinides at the RFETS FY99 report by radian
international
Viewgraphs from Jim Ranville on soil aggregation & Pu inventory

Documents and information requested for advisory group

Examples of use of RESRAD for Pu contaminated sites and decision making
Hanford for Pu (in comparison or rather than Cs and Sr and U)
NTS, LANL and other DOE Complex sites